

Tracers for Assessing Exposure to Environmental Tobacco Smoke: What Are They Tracing?

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The effectiveness of various tracers for measurements of exposure to environmental tobacco smoke (ETS) as a complex chemical mixture is based on the physicochemical properties of four major organic components and their dynamic behavior in indoor environments. For the particulate matter (PM) component and the very volatile organic compounds, emission and ventilation rates are generally the most important processes controlling indoor concentrations and exposures of nonsmokers. For the volatile organic compounds (VOCs) and semivolatile organic compounds (SVOCs), sorption on and desorption from indoor surfaces are additional processes that influence exposures. Laboratory and modeling studies of the dynamic behavior of nicotine, an SVOC, and PM indicate that nicotine can be used to estimate PM exposures from ETS in indoor environments when certain criteria are met: a) smoking occurs regularly in the environment, b) the system is near quasi-steady state, and c) sampling time is longer than the characteristic times for removal processes. Measurements in residential and workplace buildings also support the use of nicotine as a tracer for PM in ETS. Recent laboratory and field data indicate that the VOCs from ETS can be traced using compounds with similar physicochemical properties, such as 3-ethenylpyridine, pyrrole, or pyridine. The effectiveness of nicotine for estimating exposures to the VOCs and SVOCs has not been determined, although these constitute major mass fractions of ETS. — *Environ Health Perspect* 107(Suppl 2):319-327 (1999). <http://ehpnet1.niehs.nih.gov/docs/1999/Suppl-2/319-327daisey/abstract.html>

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Tobacco smoke is a complex mixture of thousands of compounds, of which approximately 400 have been measured in both mainstream smoke (MS) and sidestream smoke (SS) (1,2). Environmental tobacco smoke (ETS) is composed primarily of SS, with lesser contributions from the exhaled MS. This complex mixture of particles, gas-, and vapor-phase components is rapidly diluted and dispersed after emission and undergoes changes in its physicochemical properties because of shifts in vapor-particle distributions, sorption and desorption of vapor-phase components on indoor surfaces, and chemical

reactions. Only limited research has been conducted on these physicochemical processes, although they clearly influence the overall chemical composition of ETS and ETS exposures.

Epidemiologic studies implicate ETS exposures as a risk factor for a variety of adverse health effects in nonsmoking adults and in children (1,3,4). The nature of exposures to ETS as a complex mixture and the identification of the putative agents within this complex mixture have been addressed to only a limited extent. The epidemiologic studies generally rely on questionnaires to characterize exposures

to ETS and the available exposure measurements generally are based on determination of one or more tracers of ETS such as nicotine, as it is not possible to quantify all the constituents. Thus, two related key scientific issues are important in making ETS exposure measurements: a) How effectively do tracers measure exposures to ETS as a whole complex chemical mixture of gases, vapors, and particles? b) Does the chemical composition of ETS in different indoor environments differ significantly because of dynamic processes that remove various components at different rates and in different proportions? This article considers recent field and chamber measurements of ETS tracers and research on the various dynamic processes that affect the composition of ETS. This growing body of work has significant implications for the use of tracers to estimate exposures to the entire ETS mixture.

Understanding the dynamic behavior of ETS in indoor environments is facilitated by grouping the compounds in ETS into several major components according to their physicochemical properties such as physical state, vapor pressure, and type of compound: 1) very volatile organic compounds (VVOCs), 2) volatile organic compounds (VOCs), 3) semivolatile organic compounds (SVOCs), 4) particulate matter (PM) and its organic compounds, and 5) gas-phase inorganic compounds. Examples of the compounds in each category are given in Table 1. In this article we focus on the first four components and consider the influence of physicochemical properties and dynamic processes on estimations of indoor air concentrations and exposures.

Physicochemical Properties and Dynamic Processes Affecting ETS Components and Tracers

Exposures to ETS in indoor environments depend on smoking and ventilation rates, the volume into which the smoke is emitted and dispersed, and several other

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Abbreviations used: 3-EP, 3-ethenylpyridine; ASHRAE, American Society of Heating, Refrigerating and Air Conditioning Engineers; ETS, environmental tobacco smoke; FPM, fluorescence of a methanol extract of ETS particulate matter; MMAD, mass median aerodynamic diameter; MS, mainstream smoke; NNK, 4-(methylnitrosamino)-1-(3-pyridyl)-1-butanone; NNN, N-nitrososonornicotine; RSP, respirable particulate matter; PAH, polycyclic aromatic hydrocarbon; PDCB, p-dichlorobenzene; PM, particulate matter; RSP, respirable particulate matter; SS, sidestream smoke; SVOCs, semivolatile organic compounds; UVPM, measure of the ultraviolet absorbance of a methanol extract of ETS particulate matter; VOCs, volatile organic compounds; VVOCs, very volatile organic compounds.

Table 1. Major components of ETS classified by their physicochemical characteristics.

Major components	Vapor pressure range	Examples of compounds in the component
Very volatile organic compounds	> 7 to 13 kPa	Formaldehyde, acrolein, 1,3-butadiene, acetaldehyde
Volatile organic compounds	~0.01 to 10 kPa	Benzene, toluene, styrene, 2-butanone, phenol, pyridine, pyrrole, styrene, 3-ethenylpyridine, <i>N,N</i> -nitrosodimethylamine, <i>N</i> -nitrosopyrrolidine
Semivolatile organic compounds	10 ⁻² to 10 ⁻⁸ kPa	Nicotine, naphthalene, 1-methylnaphthalene, 2-methylnaphthalene, chrysene, fluoranthene, pyrene, <i>N</i> -nitrosomonicotine, 4-(methylnitrosamino), 1-(3-pyridyl)-1-butanone
Particulate organic compounds	< 10 ⁻⁸ kPa	Benzo[<i>a</i>]pyrene, benzo[<i>b</i>]fluoranthene, benzo[<i>k</i>]fluoranthene, solanesol
Gas-phase inorganic compounds	> 13 kPa	CO ₂ , H ₂ O, CO, NH ₃

dynamic processes such as sorption and desorption, as described in Equation 1, a time-dependent mass balance equation:

$$\frac{d(C_i V)}{dt} = E_i - QC_i - \sum_{j=1}^n S_{i,j} \left\{ k(a)_{i,j} C_i^{m(a)_{i,j}} - k(d)_{i,j} M_i^{m(d)_{i,j}} \right\} \quad [1]$$

where

- C_i = the indoor air concentration of species i from ETS, mg/m³ (other sources of i are not included here),
- V = the indoor volume, m³,
- t = time, hr,
- E_i = the emission rate for species i from ETS, mg/hr,
- Q = the ventilation rate, m³/hr,
- $S_{i,j}$ = the indoor surface area available for sorption of species i on surface j ,
- $k(a)_{i,j}$ = the deposition velocity of species i on material j , m³/hr,
- $m(a)_{i,j}$ = the adsorption rate coefficient for species i on material j , no units,
- $k(d)_{i,j}$ = the re-emission rate constant for species i from material j , per hour,
- $m(d)_{i,j}$ = the desorption rate exponential coefficient for species i , no units, and
- $M_{i,j}$ = the sorbed mass density of species i on material j , mg/m².

Thus, the concentration of any species, i , at any given time depends on the emission rate (first term of the equation) from SS and exhaled MS, the volume into which the ETS species is emitted (and convective air mixing), the rate of removal of the species by ventilation (second term), and the deposition and sorption and the desorption and

re-emission of species i from indoor surface j (third term).

Emission rates for the various constituents of ETS can be estimated as the product of the emission factors and the smoking rates in a given setting (5). Emission factors, mass of compound emitted per cigarette smoked, are important for estimating and predicting exposures to ETS over a wide range of smoking and ventilation rates and indoor conditions. Emission factors can also be used with measured concentrations of ETS tracers to estimate exposures to other ETS constituents when the tracer and constituents exhibit similar physicochemical behavior. In contrast to MS emission factors, ETS and SS emission factors generally exhibit relatively small variations across tobacco brands (6–8).

Volumes of the indoor spaces into which ETS is emitted can vary widely. The volume of a small office, for example, might be about 20 m³, whereas the volume of an industrial workplace might be of the order of several thousand cubic meters. Once emitted, ETS is diluted and mixed within the volume of the indoor space by convective mixing of indoor air. Recent experiments (9,10) have shown that the

convective mixing of indoor air is generally quite rapid, e.g., minutes in room-size spaces, particularly if there is mechanical ventilation. Mixing times correlate well with the inverse of the cube root of the power input to mechanical ventilation in accordance with theoretical predictions (10). For example, mixing time was about 7 min for a typical power input for a supply air jet. This is quite fast compared to the typical times required for removal by ventilation or for the occurrence of most adverse health effects from ETS.

Outside air ventilation rates in office buildings typically range from about 0.3 to 3 air changes per hour (11,12). American Society of Heating, Refrigerating and Air Conditioning Engineers (ASHRAE) Standard 62-1989 (13) recommends a ventilation rate for office spaces of 20 ft³/min/person. For bars and cocktail lounges, the ASHRAE-recommended design ventilation rate is 30 ft³/min per person. For retail stores, the ASHRAE ventilation rate recommendations range from 0.05 ft³/ft² of floor space (warehouses) to as high as 60 ft³/min/person for smoking lounges. For residential buildings, air exchange rates generally range from about 0.2 to 5 air changes per hour (14).

Indoor air concentrations of many ETS components are also influenced by several other dynamic processes including deposition, sorption, and desorption. The dynamic processes that affect the indoor concentrations of ETS components, the overall chemical composition of ETS, and the approximate time scales over which these occur are summarized in Table 2. The processes, which are most influential for each of the four components of ETS are discussed below in more detail.

Very Volatile Organic Compounds

The VVOCs have relatively high vapor pressures (~7–13 kPa). Their concentrations in indoor air are largely determined by emission rates, indoor volumes, and ventilation removal. In general, they are not very strongly sorbed on indoor surfaces. SS

Table 2. Dynamic processes for ETS components and their time scales.

Process	Symbol in Equation 1	Time scale
Emission	E_i	Minutes
Dilution and mixing	V	Minutes to tens of minutes
Ventilation removal	Q	Tens of minutes to hours
Deposition and sorption of vapors	$k(a)_{i,j}$, $m(a)_{i,j}$	Tens of minutes to hours
Deposition of particles	$k(a)_{i,j}$, $m(a)_{i,j}$	Hours
Re-emission of sorbed vapors	$k(d)_{i,j}$, $m(d)_{i,j}$	Hours to weeks
Diffusion into and out of solid materials	—	Days to months

emission factors for VVOCs have been summarized in the older literature by Eatough et al. (15) and Guerin et al. (2). Emission factors for some VVOCs have been measured in environmental chambers using both simulated ETS (diluted SS) (7,8) and ETS generated continuously by smokers (16). These measurements included formaldehyde, acetaldehyde, acrylonitrile, *N,N*-dimethylnitrosamine, and 1,3-butadiene, and were made using either the Kentucky reference cigarette 1R4F (University of Kentucky) or a representative sample of cigarettes from the marketplace.

Several chemically reactive VVOCs such as 1,3-butadiene and acrolein are present in high proportions in emitted SS. These species are likely to react irreversibly with indoor materials or with ozone or OH radicals in air (17,18) and thus be removed by this additional pathway. However, chemical reactions of ETS components have not been systematically investigated and the degree to which such reactions occur in indoor environments has not been determined.

Volatile Organic Compounds

VOCs generally are classified as those compounds having vapor pressures in the range of 0.01 to 10 kPa. VOCs are more sorptive than VVOCs. Emission factors have been determined for many VOCs in SS (15,19) and in smoker-generated and -simulated ETS (7,8,16,20). In addition to the influence of emission and ventilation rates on the indoor concentrations of these compounds, sorption on and desorption from indoor surfaces influence air concentrations.

Sorption and desorption of VOCs from specific indoor materials have been investigated to a limited extent. Most of these investigations were conducted in small chambers ($\leq 1 \text{ m}^3$) with higher surface-to-volume ratios than rooms (21–24). Thus, the rate constants cannot be easily extrapolated to room-size indoor environments. In general, however, both sorption and desorption appear to occur relatively rapidly, i.e., in minutes to hours.

Tichenor et al. (23) investigated the sorption and desorption of ethylbenzene on carpet, painted wallboard, and ceiling tiles in small environmental chambers at concentrations of milligrams per cubic meter. The time required to reach equilibrium between sorbed and air concentrations for these materials was about 15 hr, with an initial very rapid sorption rate over the first few hours. Sorption on all three materials was well described by the linear

portion of a Langmuir isotherm (25). After equilibrium had been reached between air and sorbed-phase ethylbenzene concentrations, fresh air was passed through the chamber to measure re-emission of the sorbed ethylbenzene. An initial rapid desorption occurred over the first few hours, followed by a longer period of tens of hours in which the ethylbenzene was more slowly emitted to yield very low air concentrations. Desorption from wallboard and ceiling tiles could be characterized using the Langmuir model (25). Re-emission from carpet deviated somewhat from this model. The data suggested that desorption from carpet is a more complex process that might involve diffusion from the solid phase (carpet backing) to the surface (fibers) followed by desorption from the surface into the room. Many other VOCs, such as benzene and toluene, would be expected to exhibit similar behavior.

There has been only limited investigation of the sorption and desorption of VOCs in room-size environmental chambers and buildings. In chamber experiments to measure emission factors for simulated ETS, Daisey et al. (7) reported evidence for the deposition and sorption of some of the higher molecular weight VOCs (phenol, the cresols, 3-ethenylpyridine (3-EP), and *N*-nitrosopyrrolidine) onto the stainless steel walls of the chamber. The rates of deposition in this 20 m^3 environmental chamber were about 0.1 to 0.2/hr. Rates in other settings will differ depending on the size of the room, air mixing, and available sorption surfaces. Desorption rates were not determined in these experiments.

Johnson et al. (26) investigated the sorption and desorption of a VOC, *p*-dichlorobenzene (PDCB), in an environmental chamber furnished with painted wallboard, carpet, and drapes and ventilated at an air exchange rate of 1 to 1.3/hr. The first set of three experiments was conducted with wallboard only; the next three experiments included carpet, and finally, three experiments were done with all three materials in the chamber. Within a few hours a steady-state concentration of PDCB was achieved due to sorption. After steady state was achieved, the emission source was removed and desorption monitored over the next 10 to 20 hr. Desorption also occurred relatively rapidly over the first hour or two, with the air concentration reaching a level of about 10 to 15% of the original steady state. In general, the monitoring data agreed well with the model that included one sink for each of

the materials. However, there was some evidence for a possible additional, slower sink for re-emission from the drapes.

Tichenor et al. (23) reported an experiment with PDCB emissions in a test house with an air exchange rate of 0.35/hr. The PDCB solid was placed in a closet in one of the three bedrooms for 11 days and air concentrations were measured on days 4, 6, 8, and 11. Air concentrations showed only small variations ($\leq 45\%$ of the lowest measured concentration) among the three rooms or over time ($\leq 30\%$) during this period. The PDCB source was removed on day 11 and air concentrations were measured on days 12, 14, 16, and 18. The air concentrations on day 12 dropped to about half those of day 11, and on day 14 to about one-fourth those of day 11, indicating re-emission of sorbed PDCB. The authors estimated that approximately 40% of the emitted PDCB had been sorbed on indoor surfaces. By day 18, air concentrations were about 10 to 15% of the original steady-state concentrations.

Semivolatile Organic Compounds

SVOCs generally are defined as those compounds with vapor pressures ranging from 10^{-2} to 10^{-8} kPa. Eatough et al. (14) summarized emission factors for many of the SVOCs in SS. Chortyk and Schlotzhauer (19) also reported SS emission factors for a number of SVOCs for 20 different cigarette brands with mainstream tar values ranging from 1 to 23 mg/cigarette. ETS emission factors for cigarette brands currently in the marketplace or for the Kentucky reference cigarette 1R4F have been determined (or can be inferred from measurements made in chambers) for nicotine (6,16,20) and some of the polycyclic aromatic hydrocarbons (PAHs) (27).

SVOCs are found in both the particle and vapor phases. Distribution depends on the temperature and age of the ETS and the particle concentration. The gas-particle distribution of nicotine has been measured by several investigators (20,27–29); most of the nicotine was found in the vapor phase. Gundel et al. (30) measured the gas-particle phase distributions of some of the PAHs in ETS using a sampler designed to minimize changes in the vapor-particle partitioning during sampling. In general, as molecular weight increased, the fraction of PAH in the particulate phase increased. Liang and Pankow (29) determined gas-particle partition coefficients for a number of SVOCs in ETS based on the desorption of the compounds from particles

collected on a filter. The SVOCs included some alkanes (C_{16} – C_{22}), several PAHs, quinoline, isoquinoline, carbazole, and nicotine. As thermodynamic consideration predicts, vapor pressure and temperature are the major determinants of the partition coefficients for the alkanes, PAH, and carbazole. For nicotine, however, the pH of ETS is a major determinant of the partition coefficient.

N-Nitrosornicotine (NNN) and 4-(methylnitrosamino)-1-(3-pyridyl)-1-butanone (NNK) are probably SVOCs also, based on their vapor pressures, but the gas-particle distribution for these and for other SVOCs have not yet been experimentally determined. Evaporative losses of VOCs and SVOCs from PM into the gas phase can also occur over time (31–33), but this appears to be a slower process than deposition to indoor surfaces.

For the SVOCs, sorption and desorption from indoor surfaces also can be significant processes that control the air concentrations. The dynamic behavior of nicotine is of particular interest, as it has so commonly been used as a tracer of ETS. Van Loy et al. (34–36) investigated the deposition and re-emission of vapor-phase nicotine in a 20-m³ stainless steel environmental chamber. Experiments were conducted first in an empty chamber in which all deposition losses were to the walls, ceiling, and floor, and then in the same chamber with a carpeted floor. Nicotine was repeatedly volatilized into the unventilated environmental chamber (the air exchange rate due to infiltration was 0.15/hr) and its decay over time was measured until a steady-state concentration was reached. This generally required 2 to 3 days following each injection. However, about 80 to 90% of the nicotine was deposited and sorbed on the surfaces within the first 1 to 2 hr after the nicotine was emitted into the chamber. The mass sorbed on the walls and carpet in equilibrium with the air was also determined. The carpet sorbed approximately 100 times more nicotine per square meter than did the stainless steel walls. After the fifth injection and decay process, the chamber was actively ventilated and resealed and the re-emission of sorbed nicotine into air monitored.

The sorption behavior of nicotine in the carpeted chamber was modeled using two different sorption dynamics models, one based on surface sorption and one based on sorbate diffusion in a homogeneous polymer (35). A deposition velocity of 4.5 m/hr and a re-emission rate constant of 0.0008/hr

were estimated for carpet from these experiments (Equation 1). In the first 5 to 6 hr after emission of nicotine into the chamber, the surface-sorption model more accurately captured the dynamic behavior of the nicotine. Over the next several days, however, the air concentration of nicotine decreased more slowly in a nearly linear fashion. This suggests that there may be two sinks operating, one rapid and surface-dominated and a second slower sink controlled by diffusion into the polymer backing of the carpet. Once there was a large sorbed mass of nicotine, reduction of the air concentration through ventilation led to a relatively rapid re-emission of nicotine into air to re-establish the equilibrium concentration.

Dynamic changes in the vapor-phase concentrations of several PAHs in ETS have also been examined over a 3-hr period in a 30-m³ environmental chamber under static conditions (no ventilation) (33). Deposition losses of the vapor-phase PAH were highly dependent on vapor pressure—more rapid losses were observed for the higher molecular weight PAH. For example, phenanthrene, pyrene, and benz[*a*]anthracene decayed faster in their gas-phase concentrations than the more volatile species such as naphthalene and its methyl derivatives. The effect of these differential deposition rates was to change the relative proportions of the different vapor-phase PAH in ETS and therefore the overall chemical composition of ETS.

Particulate Matter from ETS

Exposures to the PM component of ETS have been a major focus of much of the research on ETS. ETS emission factors for PM have been reported by a number of investigators. These range over about a factor of two, from an average of 17 mg/cigarette reported by Leaderer and Hammond (6) for 10 U.S. cigarette brands to 8 mg/cigarette reported by Daisey et al. (7) for simulated ETS (diluted and aged SS) from six commercial U.S. brands. Differences in brands smoked accounted for only about 15 to 20% of the variability (6,7). Two other factors can contribute to differences in the reported emission factors: whether the ETS included exhaled MS and experimental protocol differences that result in dynamic changes in the ETS. The higher PM emission factor reported by Leaderer and Hammond (6) was based on measurements of ETS generated continuously by smokers in a ventilated chamber with high rates of recirculated air. This "fresh" ETS

included exhaled MS as well as diluted SS. Exhaled MS accounts for 15% of ETS PM (37), which would account for some of the differences among reported emission factors. The experiments of Daisey et al. (7) involved emission, dilution, and mixing of the SS from three cigarettes into a chamber over a period of approximately 30 min. The chamber was not ventilated (infiltration was approximately 0.15/hr). The simulated ETS was held in the chamber for an additional 4 hr, with samples collected over this period, during which time there were probably volatilization losses of SVOCs and water from the PM.

Emission factors for some of the organic compounds in PM were summarized by Eatough et al. (14) and by Guerin et al. (2). More recent measurements have been reported for brands that currently are smoked in the United States and for the 1R4F cigarette. Benner et al. (31) reported concentrations of particulate nicotine, solanesol, myosmine, nicotyrine, cotinine, cholesterol, stigmaterol, campesterol, and β -sitosterol in ETS particles. Emission factors can be inferred from these data based on the emission factors reported for PM from ETS. Gundel et al. (27) reported emission factors for several particulate PAHs in ETS, including benzo[*a*]pyrene.

The mass median aerodynamic diameter (MMAD) of ETS particles in room-size chambers has been determined to be approximately 0.2 μ m (38,39). Particle size distributions of several PAHs in ETS have been measured (33) using the size-selective ETS sampler developed by Hering et al. (40). The particulate PAH in ETS exhibited a bimodal distribution. In general, the highest concentrations were associated with particles with MMADs less than 0.1 μ m, with a second mode with a MMAD of about 0.6 μ m. These results suggest that lung deposition patterns and amounts differ for the PAH particulate benzo[*a*]pyrene and ETS particle mass.

Emission and ventilation rates generally have the largest influence on the indoor concentrations of PM. There are also small depositional losses of PM to indoor surfaces. For PM, the third term in Equation 1 can be considered the difference between the deposition removal term and the re-emission term (\sim zero). Xu et al. (39) reported deposition losses of PM in ETS to the walls and surfaces of a chamber at a rate of about 0.01 to 0.05/hr. They estimated that 10 hr after one cigarette is smoked, at an air exchange rate of 0.03/hr, 22% of the particles by mass are deposited

on indoor surfaces. At 0.5 air changes per hour, 6% is deposited. There also may be losses of SVOCs from the PM over time due to removal of vapor-phase SVOCs from air by deposition to surfaces. As the vapor-phase SVOCs are deposited to surfaces, particulate-phase SVOCs volatilize from particles to restore equilibrium between the two phases.

Estimating Exposures to ETS and Its Components

Because it is prohibitively expensive to measure all or even most of the constituents of ETS in field surveys of ETS exposure, exposure typically has been quantified using two approaches: *a*) mass balance modeling based on measured emission rates and the physical characteristics of the indoor environment (Equation 1), or *b*) measurements of one or several tracers of ETS to infer concentrations of other components or of ETS as a whole. Such tracers must be unique to tobacco smoke, have similar emission rates for different cigarette brands, and have proportions consistent with those of the species they are used to trace (1). To meet this last criterion, some researchers have suggested that the tracers should be similar to the ETS species they are used to trace (20,41,42).

Tracing Exposures to ETS Particulate Matter

Much of the focus on ETS exposures has been on the PM component. Nonvolatile tracers of PM include UVP, a measure of the ultraviolet absorbance of the methanol extract of particulate matter at 325 nm (43), FPM, the fluorescence of a methanol extract of PM (44), solanesol (31,45), and scopoletin (46). The latter two tracers are high molecular weight compounds believed to be specific to tobacco smoke.

Vapor-phase nicotine (and its metabolite, cotinine) generally has been the most widely used tracer for ETS PM and ETS as a whole (6,47–49). Vapor-phase nicotine is a particularly attractive ETS tracer because it is relatively easy and inexpensive to measure and it meets the general criteria for a tracer. However, its suitability as a marker has been questioned by some researchers because it exhibits decay patterns different from those of many other ETS constituents. Chamber experiments indicate that 80 to 90% of the nicotine in freshly emitted SS is deposited to surfaces within a few hours of emission (20,34,35,42). In contrast, respirable particulate matter (RSP) from ETS is

removed from indoor environments largely by ventilation, with some smaller losses through deposition to indoor surfaces (38,39). Thus, the ratios of RSP to nicotine in ETS vary substantially with changes in time and ventilation rates (42), at least over short periods.

In contrast to the findings in short-term experiments in environmental chamber studies, Leaderer and Hammond (6) found reasonably good linear correlations between 1-week measurements of RSP and nicotine ($r^2 = 0.71$) and between nicotine and the number of cigarettes smoked ($r^2 = 0.67$) in field measurements made in 96 homes. The slope of the regression line, 10.8, was consistent with the ratio of RSP to nicotine measured for ETS in an environmental chamber. These results suggest that nicotine measured over 24 hr or more can provide a reasonable estimate of exposure to ETS particles. The results also suggest a possible quasi-equilibrium for sorption and desorption of nicotine over longer time periods and indicate that nicotine measurements made over such periods might provide reasonable estimates of exposures to other physicochemical components if we had a quantitative understanding of the dynamic behavior of those components relative to nicotine.

Van Loy et al. (36) recently examined the apparent discrepancy between short-term measurements in environmental chambers and the longer term nicotine and RSP measurements made in residential buildings. They used new models to examine the effects of reversible sorption on nicotine's suitability as an ETS PM marker. The dynamic behaviors of nicotine and PM from ETS were modeled for both the environmental chamber experiments described by Nelson et al. (42) and for 24-hr average concentrations in a 500-m³ house in which smoking occurs regularly for 16 hr/day. Results showed that the apparently contradictory observations could be reconciled by taking into account the sampling times and the dynamic behavior of nicotine, including its sorption and re-emission under different environmental conditions. Specifically, in indoor environments in which smoking occurs on a fairly regular basis for an extended period, for example, the 96 homes in which Leaderer and Hammond made measurements, the sorbed mass of nicotine becomes large relative to the mass emitted by a single cigarette, and re-emission from indoor surfaces becomes significant relative to direct emission. Furthermore, the time intervals during which air concentrations

are elevated (immediately after nicotine emission) are quite short relative to the sampling period. Thus, the average air concentration of nicotine remains relatively constant and the ratio of nicotine to PM is observed to be relatively constant. The environmental chamber experiments of Nelson et al. (42) and others are examples of conditions under which the ratio of nicotine to PM changes rapidly because of the rapid deposition to surfaces that occurs immediately after emission. The shorter measurement intervals spanned periods in which the air concentration of nicotine changed dramatically.

Evidence from field measurements supports the idea of indoor reservoirs of sorbed nicotine that are reemitted into air when smoking has stopped. Vaughan and Hammond (50) measured average weekly nicotine concentrations in offices of smokers that ranged from 4 to 24 µg/m³ before a smoking ban. Seven weeks after the ban, nicotine was still measured in the air but at concentrations ranging from 0.1 to 0.5 µg/m³. This suggests a large remaining reservoir of sorbed nicotine.

The recent personal monitoring measurements reported by Jenkins et al. (49) also indicate that nicotine is a suitable tracer for the PM from ETS. These investigators measured various tracers of ETS for 24-hr periods for four groups: smokers working in environments in which smoking was permitted, smokers working in nonsmoking environments, nonsmokers working in workplaces in which smoking was permitted, and nonsmokers working in nonsmoking workplaces. Figure 1 shows the relationship between the percentile concentrations of nicotine and PM reported by Jenkins et al. (49) for the different groups. The relationship is linear ($r^2 = 0.91$) and the slope of the line, 10.9, is in good agreement with the slope of the

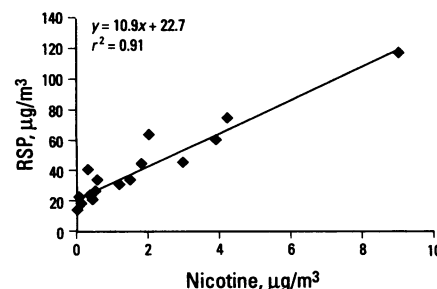


Figure 1. Linear regression of RSP versus nicotine for data reported by Jenkins et al. (49).

regression line between RSP and nicotine for 96 residences in New York State (6).

Jenkins et al. (49) also measured the ETS particulate tracer solanesol. The correlation coefficient for the regression of solanesol with RSP for the 12 data points from groups 1, 2, and 3 (for which all data were above the limit of detection) is 0.92, and the slope of the regression line, which should be equivalent to the ratio of solanesol to RSP from ETS, is 0.018. Benner et al. (31) reported that the concentration of solanesol in ETS particulate matter is 22.2 mol/g or 0.017 g/g of PM. This agrees well with the ratio implied by the data of Jenkins et al. (49) and supports the use of solanesol as a tracer of PM from ETS. A similar regression of concentrations of scopoletin versus RSP for the Jenkins et al. data yields a regression line with more scatter ($r^2=0.90$)—probably because of the difficulty of the scopoletin analysis—and a slope of 0.69 ng scopoletin per μg RSP. Risner (46) reported an emission factor of 111 ng scopoletin/cigarette that combined with an RSP emission factor of 17 mg/cigarette (6) implies a concentration of 0.0065 ng scopoletin/ μg PM from ETS. The reason for this discrepancy is not known.

Estimating Exposures to VOCs and SVOCs from ETS

VOCs and SVOCs constitute a major proportion of the organic mass of ETS, and many of the compounds in these fractions are known to be biologically active as carcinogens in animals. Gas-phase ETS also has been implicated in coronary heart disease (51). However, efforts to trace exposures to these constituents have been much more limited than those for PM. There

have been only a few investigations of the contributions of ETS to VOC exposures in offices. Comparisons of the concentrations of selected VOCs in smoking and non-smoking offices have been reported by Bayer and Black (52) and by Proctor et al. (53). They found little difference in VOC concentrations that could be attributed to smoking. However, estimating the ETS contribution to VOCs is complicated by the differences among buildings in the relative contributions of such sources and in ventilation rates. Thus, it is unlikely that any consistent differences would be observed, even when smokers contributed to the indoor concentrations of these compounds.

Therefore, ETS tracers or mathematical models based on the mass balance principle are needed to provide estimates of the ETS-attributable exposures to VOCs. Daisey et al. (54) did this for four office building scenarios based on a mass balance model and emission factors for VOCs in SS available in 1990. Table 3 presents the modeled concentrations for one VOC and three VOCs for the two of the cases considered in Daisey's paper (54) but with ETS emission factors (7). For comparison Table 3 also includes the concentrations of these compounds found in office buildings in which smoking was allowed (52,53).

Several VOCs unique to ETS have been examined as possible tracers of other VOCs in ETS. Heavner et al. (55) investigated the use of 3-EP as a tracer of VOCs in ETS in residential settings. This compound was not found in homes of nonsmokers (55) and there was a significant correlation between 3-EP and smoking activity in homes of smokers. Heavner et al. (55) used source apportionment based on 3-EP and

the ratio of 3-EP to benzene and styrene in ETS to estimate contributions of benzene and styrene from ETS in the homes.

Hodgson et al. (56) evaluated the use of three VOC tracers for ETS. These investigators compared measured to modeled concentrations of pyridine, 3-EP, and pyrrole in smoking rooms of office buildings. Emission factors determined in an environmental chamber (7) were used in a mass balance model to estimate indoor concentrations of the tracers in the smoking lounges. The measured and modeled concentrations in the smoking lounges were in good agreement for 3-EP and pyrrole, and this result validates the use of these two tracers. Table 3 presents the estimated ETS contributions to formaldehyde and three VOCs in the smoking rooms of offices based on the concentrations of the 3-EP tracer. The estimated indoor air concentrations of these compounds from ETS in the smoking rooms are consistent with the modeled concentrations for Case 4 (54) for offices in which smoking is allowed. Hodgson et al. (56) estimated that ETS contributed from 20 to 84% of these four VOCs in the smoking rooms. Fractional contributions of ETS to acetone, toluene, ethylbenzene, xylene isomers, and α -limonene were all less than 50%.

Unfortunately, Hodgson et al. (56) did not examine the relationships between nicotine and the VOCs in these office buildings. However, linear regression analysis of the concentrations of nicotine and 3-EP tracers reported by Jenkins et al. (49) can provide some information. Figure 2 shows the high correlation between these two tracers. The slope of the linear regression line, which is the ratio of 3-EP to nicotine, is 0.4. This is considerably lower than the ratio of 0.7, which can be calculated from environmental chamber measurements (7). This difference in the ratio may be due to the difference between the

Table 3. Measured concentrations and estimated ETS contributions of selected VOCs in office buildings in which smoking is permitted.

	Concentrations, mg/m ³			
	Formaldehyde	Benzene	Styrene	2-Butanone
Measured				
Bayer and Black (53), totals from all sources, measured in offices of smokers	—	ND–6.0	—	—
Proctor et al. (54), total from all sources, measured in smokers offices	—	4–8	4–15	—
Modeled				
ETS contributions to VOCs, Case 2 ^{a,b}	18.9	5.8	2.1	4.2
ETS contributions to VOCs, Case 4 ^{a,c}	8.2	2.5	1.0	1.8
3-EP tracer estimates				
ETS contributions to VOCs based on 3-EP tracer measurements in smoking rooms ^d	0.9–5.8	1.3–8.2	0.5–3.0	1.3–8.2

ND, not detected. ^aDaisey et al. (54), with updated emission factors taken from Daisey et al. (7). ^bCase 2: two cigarettes per hour per smoker, 20% smokers, an occupancy of 23.8 m² per worker, 0.47 air changes per hour during the workday (ASHRAE standard) and 0.41 air changes per hour during nonworking hours. ^cCase 4: two cigarettes smoked per hour per smoker, 20% smokers, an occupancy of 23.8 m² per person, and a daytime air exchange rate of 1.27 air changes per hour during the workday and 0.41 during nonworking hours. ^dData from Hodgson et al. (56).

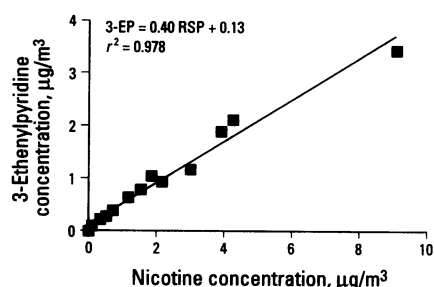


Figure 2. Linear regression of 3-ethenylpyridine versus nicotine. Data from Jenkins et al. (49).

sorption characteristics of a real room with sorptive furnishings and the unfurnished environmental chamber.

There are many other VOCs and SVOCs that are of interest, for example, the *N*-nitrosamines. Presently we lack experimental evidence that they can be traced effectively by nicotine or even by a VOC tracer such as 3-EP or pyrrole. Simultaneous measurements of multiple VOCs, SVOCs, and the various tracers in environmental chambers under more realistic conditions (with furnishings, repeated cycles of smoking, etc.) and in real buildings are needed to determine the relative proportions of these components to ETS tracers. Such experiments are also needed to evaluate the accuracy of various ETS tracers in tracing exposures to the entire ETS mixture.

Implications of Existing Data on the Chemical Composition of ETS

It has generally been assumed that the overall chemical composition of ETS is similar to that of diluted SS. There are sufficient data to indicate that this assumption is incorrect for at least some of the VOCs and SVOCs in ETS that are sorbed on and desorbed from indoor surfaces. For example, the emission factor for nicotine in SS is about 5 mg/cigarette (7,57), whereas the emission factor measured in environmental chambers in which dynamic processes occur is only about 1 mg/cigarette. It is interesting to note that the emission factor calculated from the measurements by Eatough et al. (20) of gas-phase nicotine in a Teflon-walled environmental chamber is more consistent with that of SS, presumably because nicotine does not sorb on Teflon. There have been only a few experiments on sorption and desorption of VOCs and SVOCs in environmental chambers furnished to simulate real indoor environments (58). Such experiments are needed to better characterize the relative proportions of these compounds in ETS and their dynamic behavior and to evaluate the effectiveness of various ETS tracers. These experiments can also provide some insight into one of the questions raised in the introduction, i.e., "Does the chemical composition of ETS in different indoor environments differ significantly because of dynamic processes that remove different components at different rates and in different proportions?"

Summary and Conclusions

This paper has considered the dynamic behavior of four major physicochemical components of ETS and recent experimental evidence on the effectiveness of various tracers for measuring exposures to ETS as a whole and to major components of ETS in indoor environments. For PM and the unreactive VVOC components, emission and ventilation rates are probably the most important processes controlling indoor concentrations and exposures of nonsmokers. For the VOCs and SVOCs, sorption on and desorption from indoor surfaces are also significant processes controlling the indoor concentrations. There is evidence that the distributions of gas- and sorbed-phase compounds in these two groups are functions of vapor pressure and molecular weight, with a higher proportion of higher molecular weight compounds sorbed on surfaces. However, the behavior of these ETS compounds has not been sufficiently investigated to allow their concentrations to be predicted with any confidence.

Field measurements in residential and office environments (6,49) indicate that nicotine is a good tracer for PM. Laboratory and modeling studies suggest that nicotine measurements can be used to estimate PM from ETS in indoor environments when the following criteria are met: *a*) smoking occurs regularly in the environment, *b*) the system is almost quasi-steady state, and *c*) sampling time is significantly longer than the characteristic times for removal processes.

When these criteria are not met, for example, for certain individual residences or offices, nicotine is probably a less reliable quantitative indicator of PM exposures from ETS, although how biased a measure it provides in such circumstances remains to be determined.

The field measurements of Jenkins et al. (49) combined with laboratory measurements support the usefulness of a particulate tracer such as solanesol for tracing PM from ETS. Because there is reason to believe that the dynamic behavior of the VVOCs is dominated by the same factors that dominate PM, tracers for PM may also be good tracers for unreactive VVOCs from ETS.

VOCs and SVOCs also constitute a significant proportion of total ETS mass. Recent laboratory and field data (56) provide evidence that VOCs from ETS can be traced using compounds within this component such as 3-EP, pyrrole, or pyridine,

which have similar physicochemical properties. Source apportionment of VOCs based on field measurements of 3-EP and the ratio of 3-EP to the VOCs indicates that ETS can be a significant contributor to the total VOCs in office smoking rooms (56). To date, no systematic investigations have been made of the effectiveness of any of the usual ETS tracers for estimating exposures to the SVOCs in ETS.

Nicotine may also prove to be a useful tracer for the VOCs and SVOCs from ETS if the previously mentioned criteria are met. However, there are significant differences between the ratios of 3-EP and nicotine measured in the field and in an unfurnished environmental chamber. This is hypothesized to be because of differences in the sorptive materials in these two environments. If this hypothesis applies to other VOCs and SVOCs, the ratios of the compounds to nicotine (and other tracers) would have to be determined in furnished environmental chambers using more realistic smoking patterns and ventilation rates.

It is clear from existing data that freshly generated and diluted SS is not chemically identical to ETS, but we do not yet have quantitative information on the relative proportions of many of the VOCs and SVOCs in ETS. Carefully designed experiments should be conducted in environmental chambers furnished to better simulate real indoor environments to provide this information. Such experiments should also provide much needed experimental evidence that nicotine can be used to trace the VVOCs, VOCs, and SVOCs in ETS under some conditions.

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